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LLNL HEPA Filter Support of Nuclear Facility Safety R&D (FY12)

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Executive Summary

The ceramic HEPA filter project has been a successful and innovative project. This report contains information from the work from FY12 funding, as well as remaining FY11 funding. Of note, a total of 12 patents for the ceramic HEPA filter program have been filed, and an additional record of invention has been prepared with a provisional patent to follow. This project supports development of a ceramic HEPA filter technology to benefit DOE nuclear facilities by providing lower life-cycle costs and reducing or eliminating costs associated with Safety Class and Safety Significant systems in nuclear facilities. The satisfactory performance of ceramic filters in a fire could significantly reduce costs of support systems associated with mitigating a release, such as fire suppression, fire detection and alarm, and internal building structure. In nuclear safety, the best way to provide protection to public, workers and the environment is to provide a barrier between the hazard and the receptors. A passive engineered design feature is the best barrier for that purpose. Advanced ceramic HEPA filters provide an excellent barrier between radioactive material in a nuclear facility and everything downwind.

Both university (Cal Poly) student teams did very well. As summarized in this report, they successfully added and upgraded instrumentation and controls to the High Temperature Test Unit (HTTU) for the ceramic filter project. They also succeeded in adding viewing ports, remotely controlled closed circuit television, a flame impingement system and a helium filter gasket leak detection system. Details can be found in Ref. 3 - 6. The HTTU has flexible capabilities and can also be used to test other types of filters or filter components in the future, if so desired.

At LLNL, research to improve filter media coating (Fig. 1) is being conducted to maintain filtration efficiency while improving performance (reducing pressure drop). Improvements in the individual filter element are necessary to maintain filtration efficiency while decreasing pressure drop. The path forward for an optimal ceramic HEPA filter meeting DOE's needs is to improve the filtration media. This task utilized the LLNL/DOE developed innovations along with progress in previous activities to prepare multiple ceramic filter elements and refine the technical details for depositing advanced filtration materials to the ceramic filter to meet desired filter performance. The ceramics filter elements were tested for pressure drop at the required air flow rate using an air flow test system. The filtration efficiency and pressure drop of the ceramic filter elements will be measured as part of the development process. Details can be found in Attachment A.

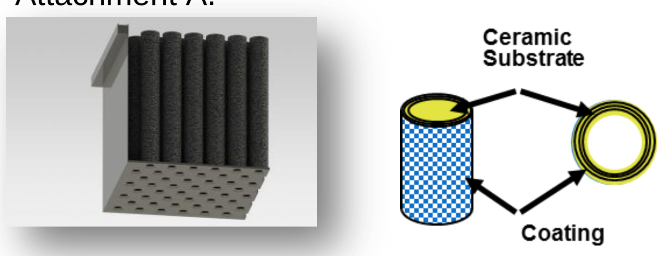


Figure 1. Conceptual diagram of ceramic filter and filter element

Task 1. Testing ceramic HEPA filter(s) at ICET.

The ability to test ceramic HEPA filters is important to the overall project of developing ceramic HEPA filters. LLNL discussed options with ICET regarding the optimal ICET test equipment to use. ICET's metal test stand (used for qualification of metal HEPA filters for ASME AG-1 Section FI (metal filters)) was selected as the best test equipment to test ceramic HEPA filters.[1] See Figure 2. The same test stand will be used to support a new AG-1 Subsection (FO) for ceramic HEPA filters.[2] It was determined that LLNL should wait until after ICET has modified the test stand for ceramic filters and has developed the test plan for AG-1 Section FO.[3] It was also determined that the proof-of-concept filter design could be later tested at ICET, but not the first proof-of-concept filter itself due to geometric constraints. LLNL interfaced with ICET to ensure that LLNL produces and ships to ICET a filter assembly that meets their requirements. If necessary, there may be higher temperature testing at LLNL to debug potential issues prior to testing at ICET. ICET sent LLNL a blank mounting plate which LLNL is modifying to accommodate ceramic test filters.

Figure 2. Selected test stand at ICET

Deliverable: Report on the project efforts and accomplishments (this document).

[1] During this period, ICET conducted AG-1 Section FI shakedown testing on the metal test stand.
[2] LLNL interfaced with ICET to formally initiate the ASME AG-1 Section FO (for ceramic filters) writing team. The Project Initiation Form (PIF) was filed, kickoff and follow up meetings held, and a path forward established for writing Section FO.
[3] ICET's work on the development of qualification testing standards (filter qualification test setup and specifications for industry codes and standards) for ceramic HEPA filters is a separate project.

Task 2. University Collaborative effort

Part A. High temperature testing unit for HEPA filters.

In order to optimally develop ceramic HEPA filter capabilities, more than just the filter media needs to be upgraded. The frame seal and media binder need to be optimized as well. In order to develop and test new frame sealants, media binders, and other materials, and designs,[4] development of a high temperature HEPA filter testing unit is required. A review of universities and their capabilities resulted in the California Polytechnic State University, San Luis Obispo (Cal Poly) being selected to design, fabricate, test and demonstrate a High Temperature Testing Unit (HTTU) for HEPA filter assemblies and components (see Fig. 3, 4, and 5). LLNL consulted with ICET to ensure that the HTTU was developed as a compliment to ICET capabilities. The HTTU is primarily focused on ceramic filters, but can also support studies of non-ceramic filters and their materials if desired by NSR&D, DOE-EM, or NNSA. This continuing task is to provide an engineering solution to a manufacturing problem involving complex materials, material interface, and manufacturing issues. Specifically, the task is to design, fabricate, test and demonstrate enhancements to the HTTU. Cal Poly Students and faculty have continued to provide excellent work and value. Student teams have consistently under run cost estimates and provided comprehensive project reports.

Figure 3. Cal Poly Engine Laboratory, home to the HTTU

Figure 4. HTTU Testing

[4] Cal Poly's subsequent testing of sealants, gaskets, and filter media is a separate project.

Figure 5. HTTU Overview

Team CP

On May 11, 2012, LLNL and the Cal Poly Corporation agreed to Modification 1 to their contract to add controls and instrumentation to the HTTU. A team of three senior students were subsequently assigned to the project (Team CP HEPA). From January through December 2012, the students, Cal Poly faculty advisors, and LLNL collaborated on the development of HTTU controls and instrumentation (Ref. 3). The final approach met the needs of this project. On April 10, 2012, the students traveled to LLNL and presented their preliminary design to laboratory senior management and technical representative (Ref. 4). The preliminary design was validated and material acquisition begun. A final design report was completed in November 2012 (Ref. 4). See Figures see Fig. 6, 7, and 8. Additional information can be found in Ref. 3 and 4.

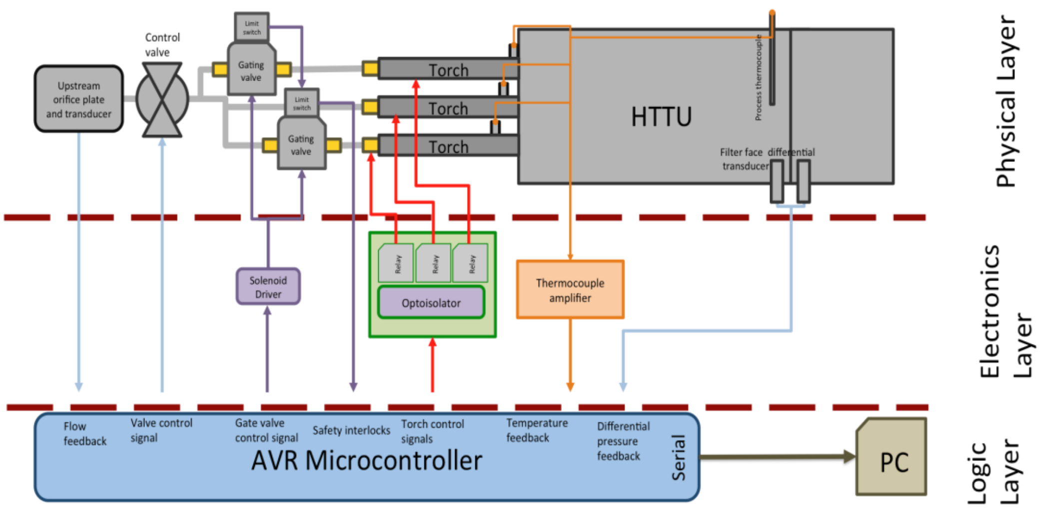


Figure 6. System Schematic

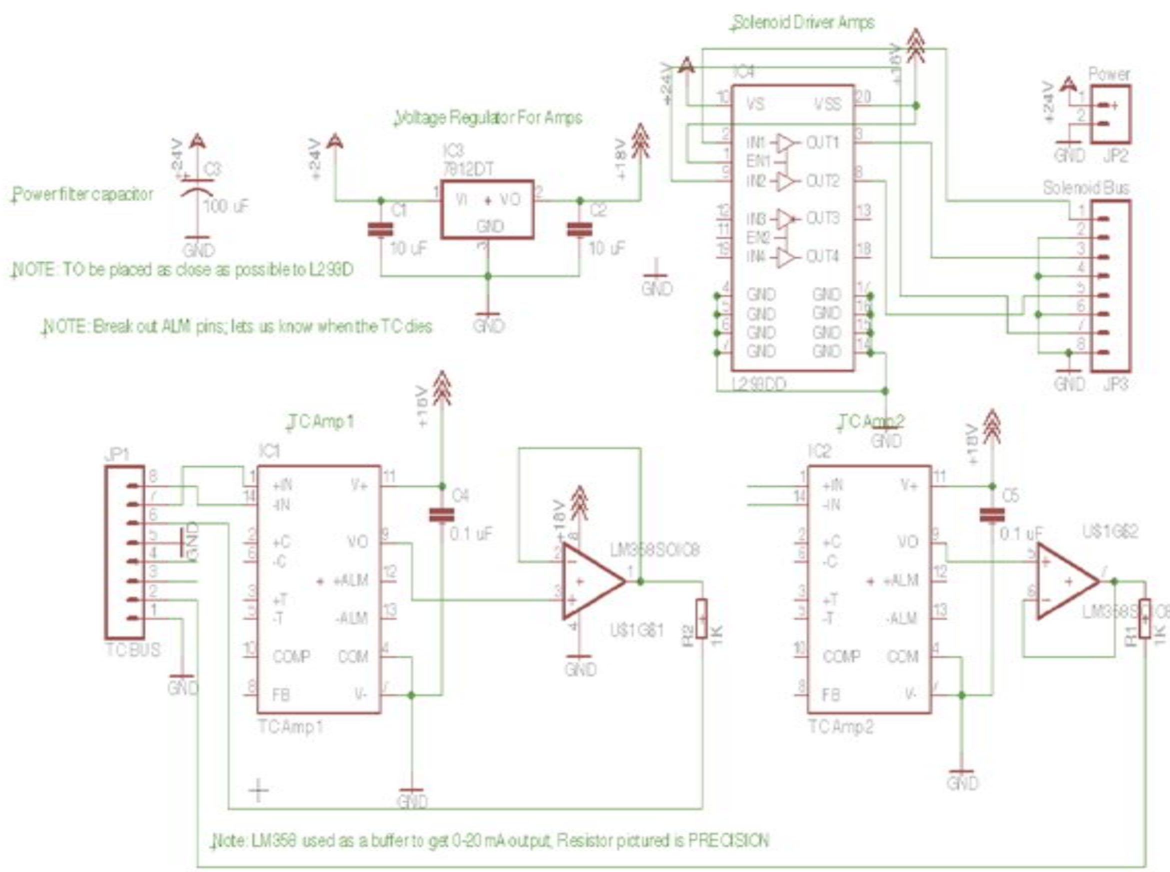
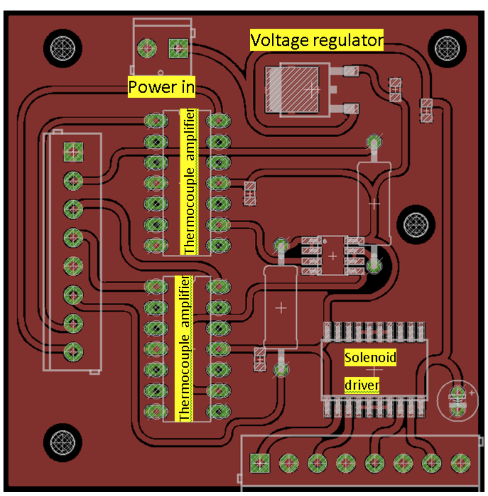
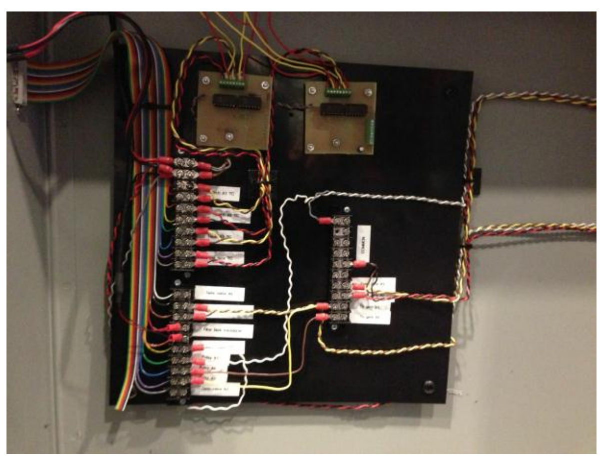


Figure 7. Thermocouple Amplifier/Solenoid Driver (TAD) Board Design



TAD Board Design Layout



TADS in HTTU Low Voltage Enclosure

Figure 8. TAD Design and installation

In mid-September 2012, Team CP HEPA completed the majority of the fabrication of the HTTU controls and instrumentation installation and successfully ran the unit. In the ensuing weeks, system characterization, sensor calibration, noise reduction, and UI testing were performed. See Figures see Fig. 9 and 10.

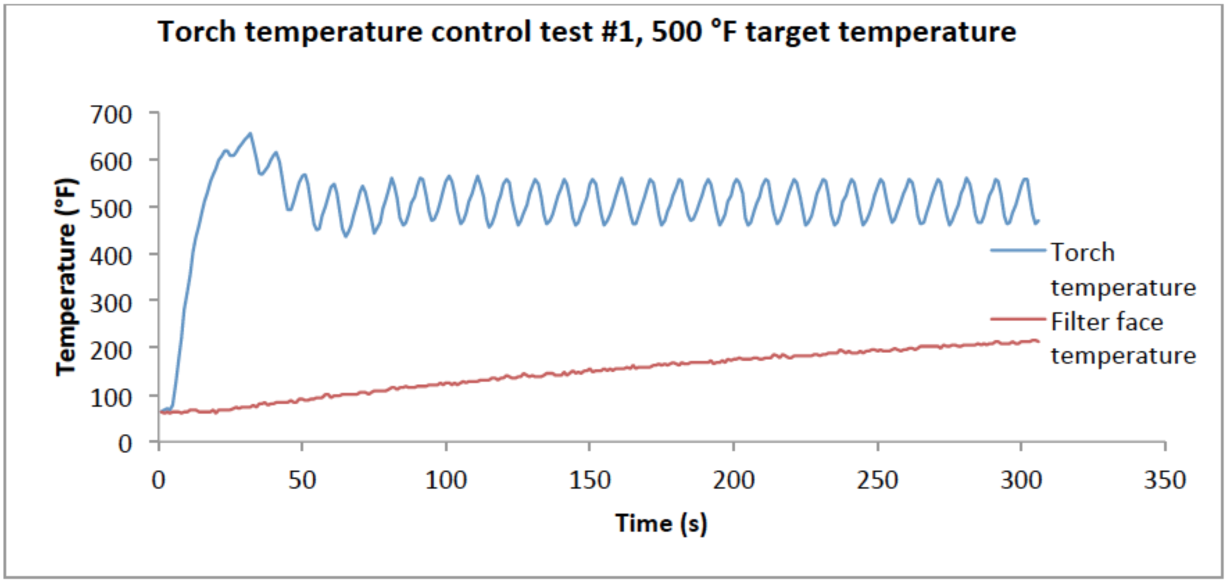


FIGURE 35 TORCH CONTROL RESPONSE, EARLY TESTING

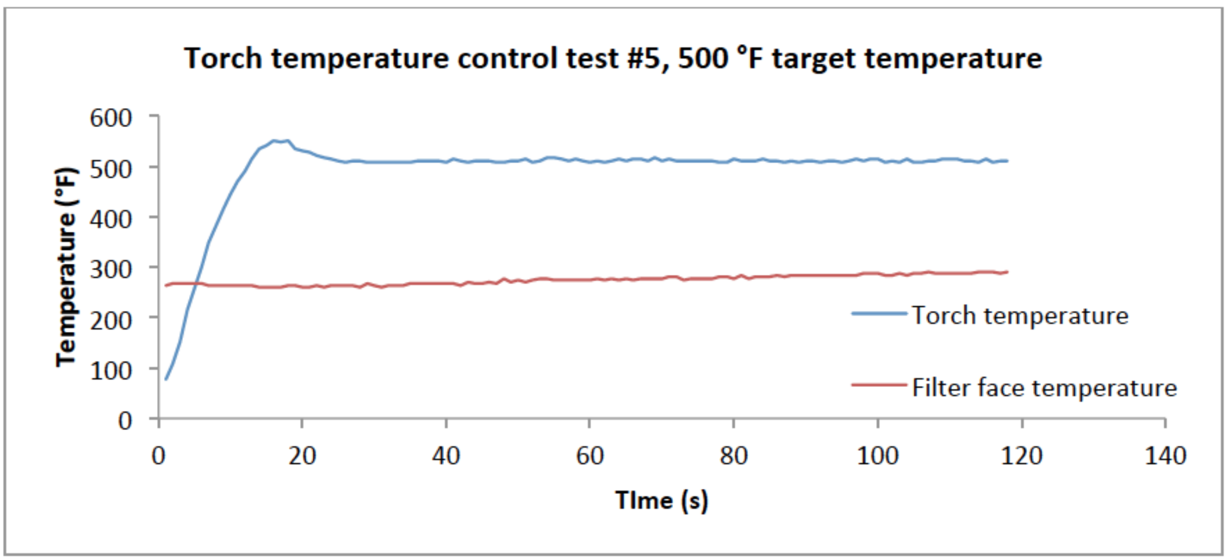


FIGURE 36 TORCH CONTROL RESPONSE, FINAL TUNING PARAMETERS

Figure 9. Torch Temperature Control Tests

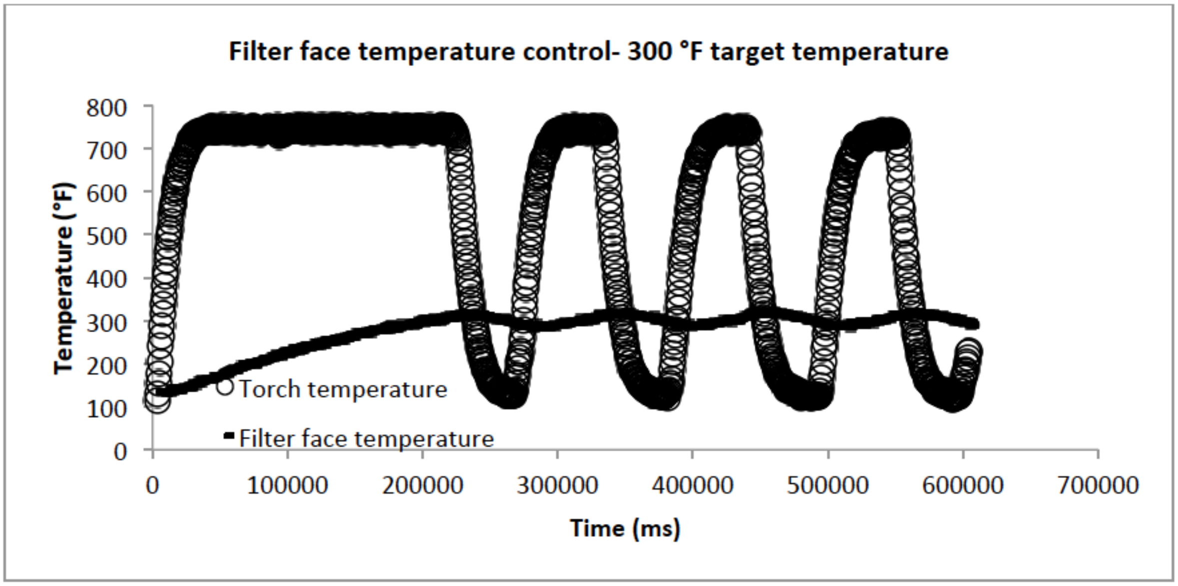


FIGURE 37 UNSTABLE OPERATION OF TORCHES AND FILTER FACE TEMPERATURE CONTROL

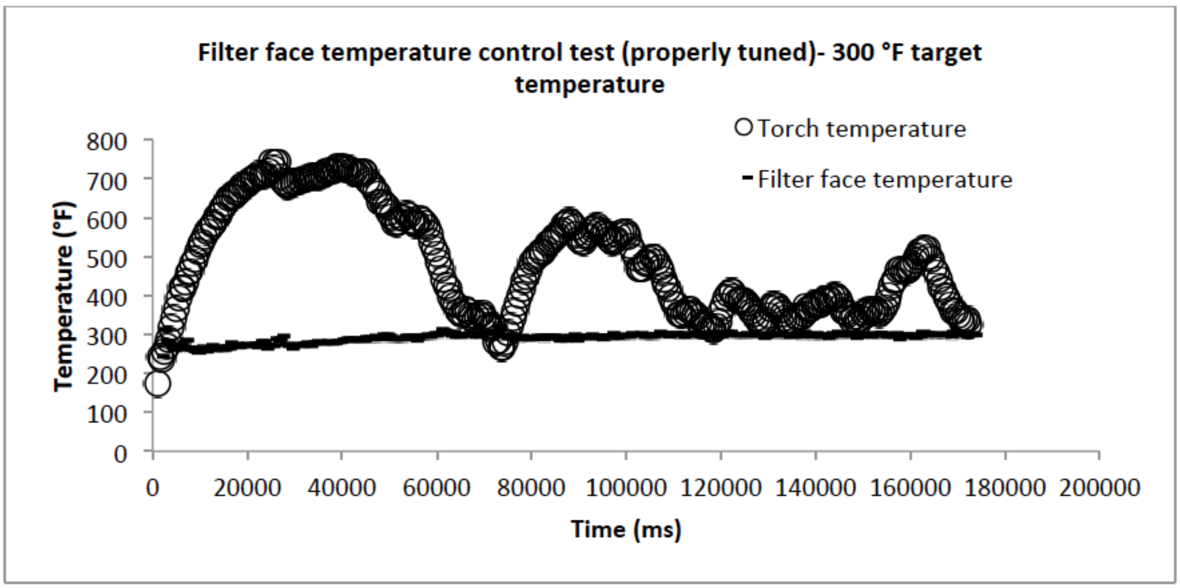


FIGURE 38 FILTER FACE TEMPERATURE CONTROL TEST, STABLE OPERATION

Figure 10. Filter Face Temperature Control Tests

CalPoly Deliverable: Team CP HEPA provided the deliverables:

1. Design drawings
2. Description of system
3. HTTU Control and Instrumentation system
4. HTTU performance test results
5. Final Design Report

LLNL Deliverable: Report on the project efforts and accomplishments (this document).

Team HiTop

On April 13, 2013, LLNL and the Cal Poly Corporation agreed to Modification 2 to their contract to add viewing ports, CCTV, a flame impingement system and a filter gasket leak detection system to the HTTU. A team of four senior students were subsequently assigned to the project (Team HiTop). From September 2012 through May 2013, the students, Cal Poly faculty advisors, and LLNL collaborated on the development of the HTTU enhancements. The final approach met the needs of this project. On February 8, 2013, the students traveled to LLNL and presented their preliminary design to laboratory senior management and technical representative (Ref. 5). A final design report was completed in June 2013 (Ref. 6). Additional information can be found in Ref. 5 and 6.

View Port

The goal of adding a view port is to provide a visual access point for the camera system in order to provide a record of tests. Team HiTop selected high temperature ceramic glass option as it was the most cost effective and comes more readily in the geometry that they needed. A stacked-frame double-pane design was chosen because meets the high temperature and limited thermal loss requirements. See Fig. 11, 12, and 13.

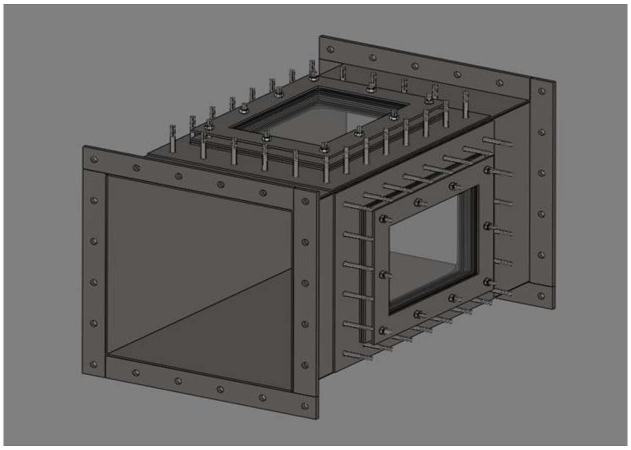


Figure 11. Stacked Frame Viewports Design

Figure 12. Prototype Viewport

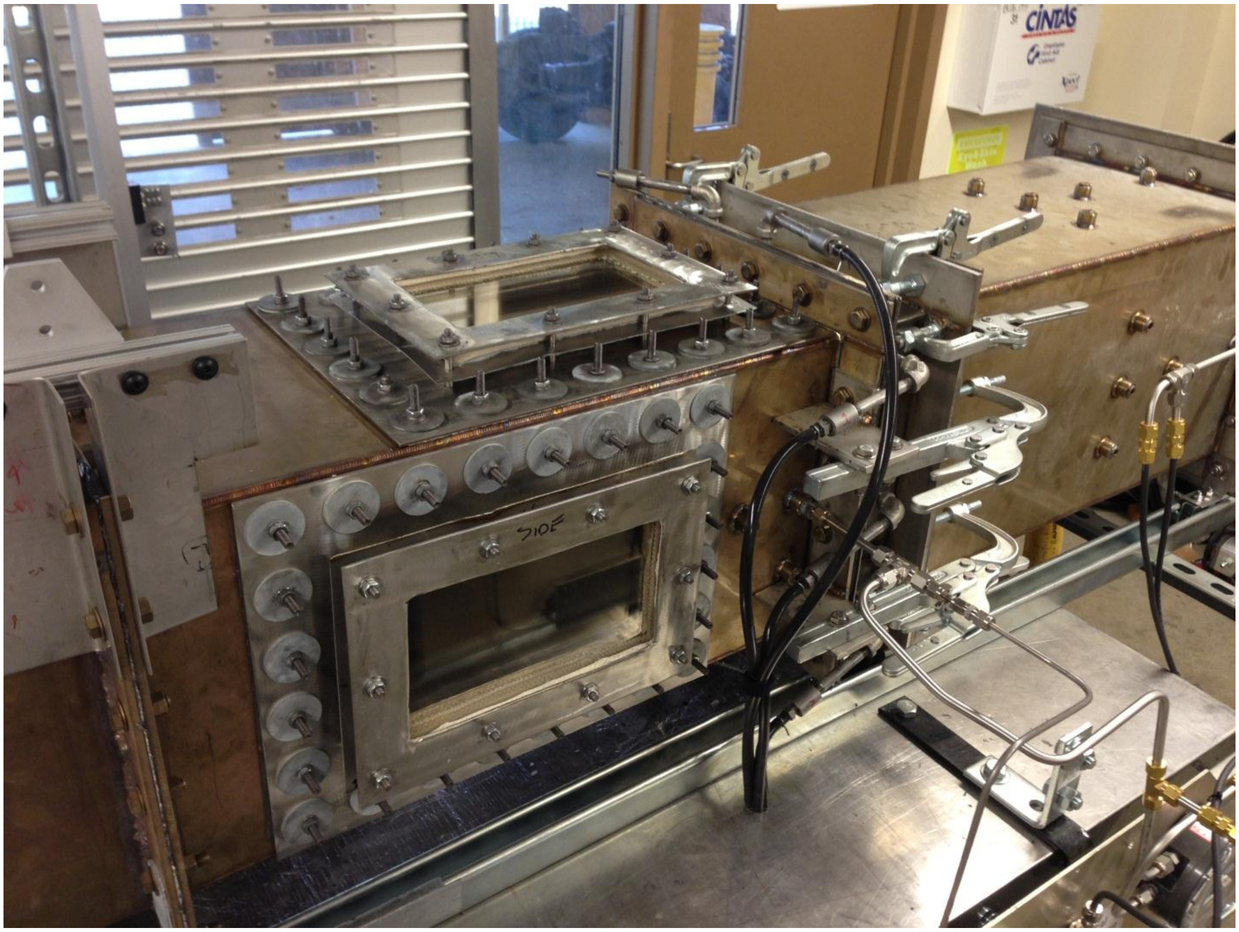


Figure 12. Final Installed Viewing Ports

Camera System

The goal when designing the frame for the camera system was to create a mount which was highly versatile, lightweight, and easy to manufacture. The following figures depicts the final design for the frame as it will be installed on the unit (see Fig. 13, 14, and 15).

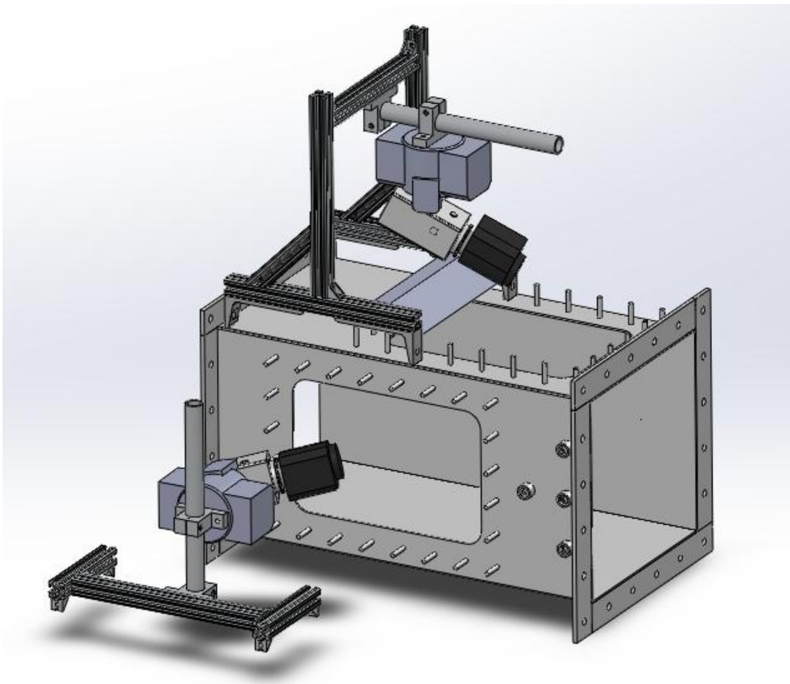


Figure 13. Camera System Frame

The basic layout for the camera network is shown below:

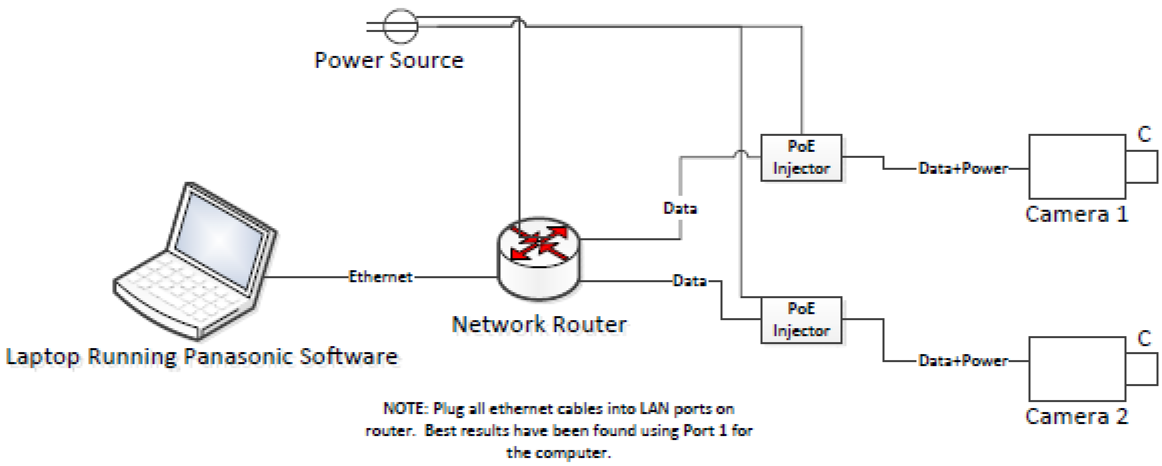


Figure 14. Network Setup of Internet Protocol Cameras

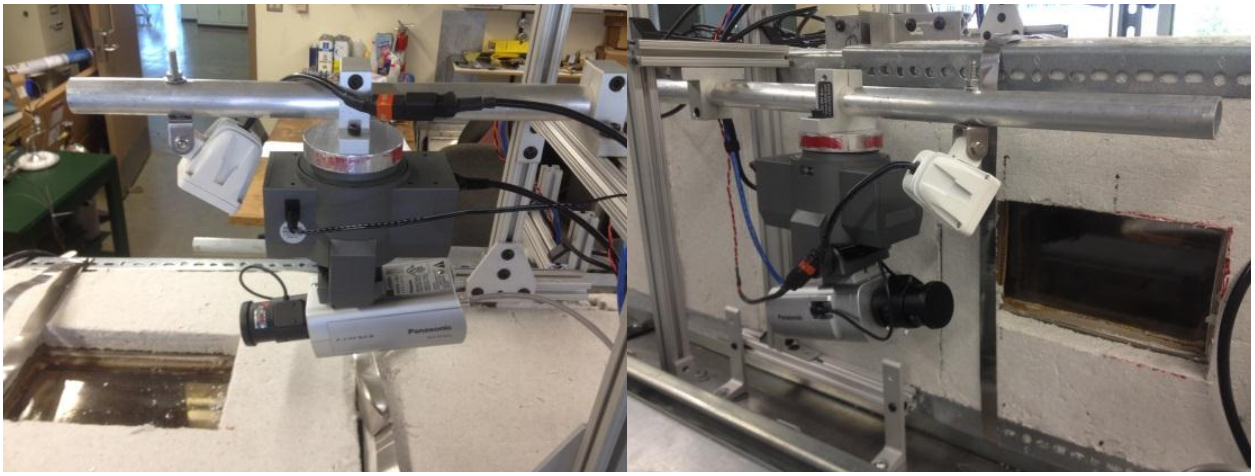


Figure 15. Final Camera Assemblies

Leak Detection

The goal for a leak detection system is to both quantify and locate leaks in the filter seals. This must be done for both the gel type seals and the gasket type seals. The helium based system uses a collector system, mounted on the outside of the filter seal, to bring air through a valve manifold and into a helium detector. See Figures 16 -25. The proposed layout for the helium-based leak detection system is shown in Figure 16 below.

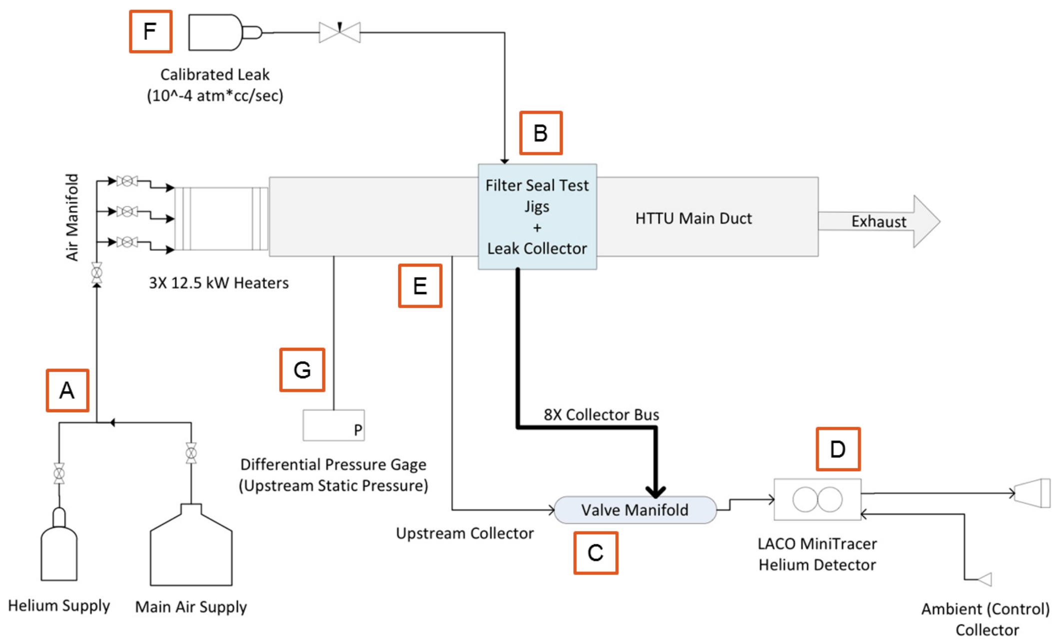


Figure 16. Proposed Leak Detection System

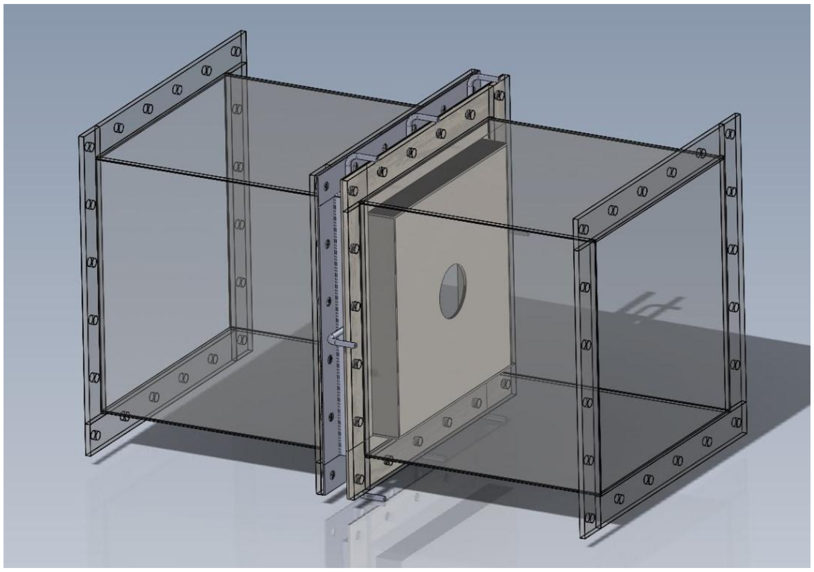


Figure 17. Isometric view of final collector design assembled between two ducts

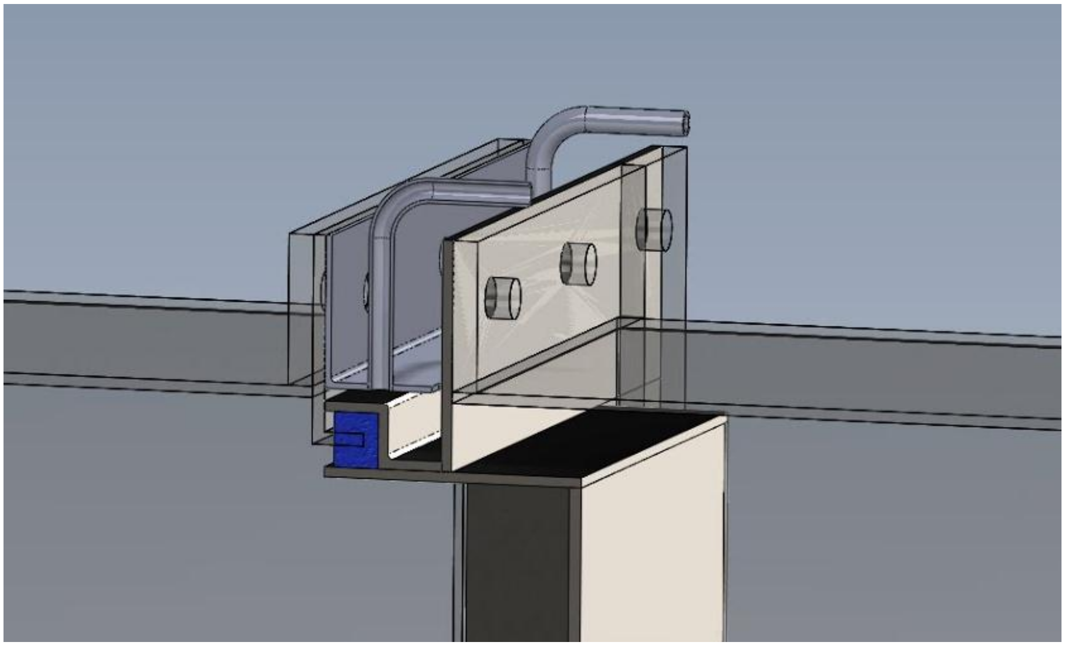


Figure 18. Cross section of collector design shows geometry of collector volumes


Figure 19. Left to right: Gasket seal test jig, collectors, gel seal test jig, in process of being welded

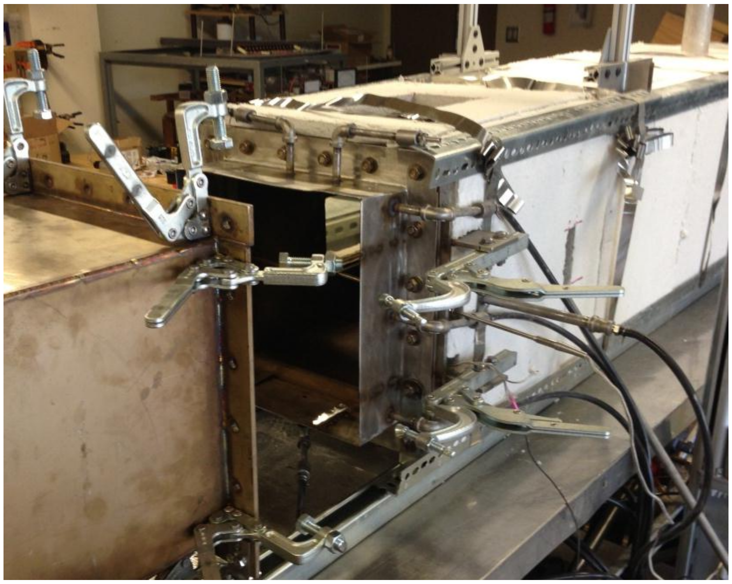


Figure 20. HTTU with module clamps open, allowing access between upstream and downstream sections.

Figure 21. HTTU with module clamps closed, applying pressure to the filter seal similar to the original filter mounting mechanism.

Figure 22. Valve manifold for leak detection system as installed in the HTTU.

Figure 23. Valve manifold control box.

Figures 20 - 23.

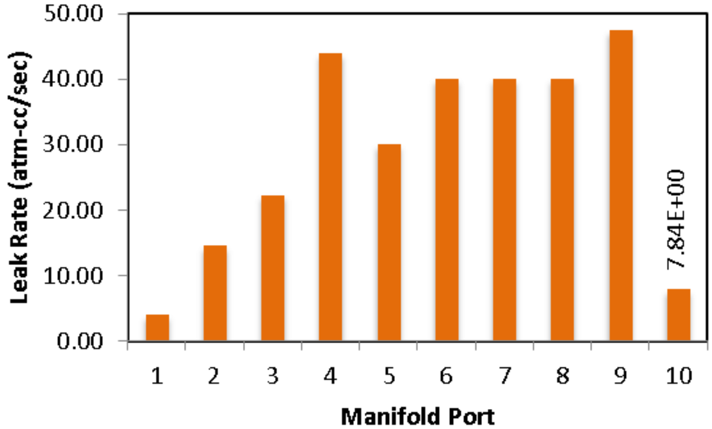


Figure 24. Graphical representation of leak rate data for first test. During this first run, not enough time was allowed between areas to for system to clear. Inability to control test port pressures resulted in high leak rates.

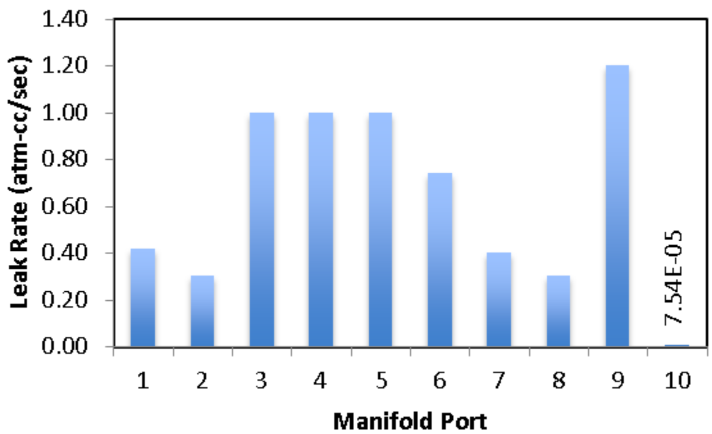


Figure 25. Graphical representation of leak rate data for second test. Note the order of magnitude with reference to Figure ; inclusion of a needle valve lowered test port pressure and provided more accurate results.

Figures 24 - 25.

Flame Impingement

The goal of the flame impingement apparatus is to test the response of a HEPA Filter to an "ember" - like flame. The method of approach for implementing the torch system design is to generate flame with gas torch. The torch is moved to impinge the flame on the desired surface; this is controlled externally. The torch system is broken into three subsystems. The motion mechanism consists of two linear actuators to give the radial and angular motion in and out of the HTTU. The gas system is propane based. The electrical system is a high temp igniter and wiring for inside the HTTU. See Fig. 26 – 29.

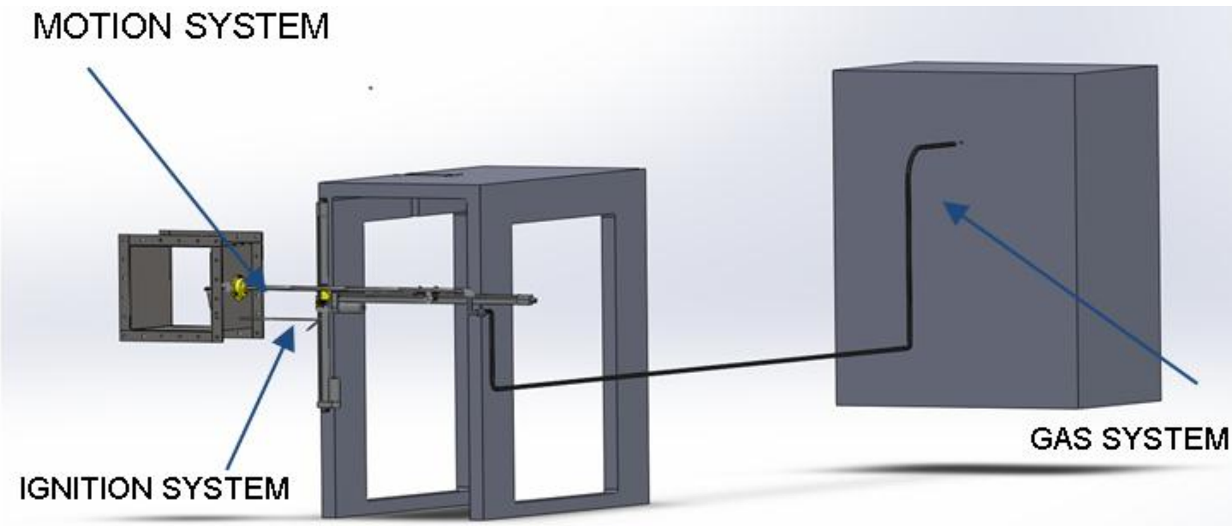


Figure 26. Torch System

Figure 27. Controllers

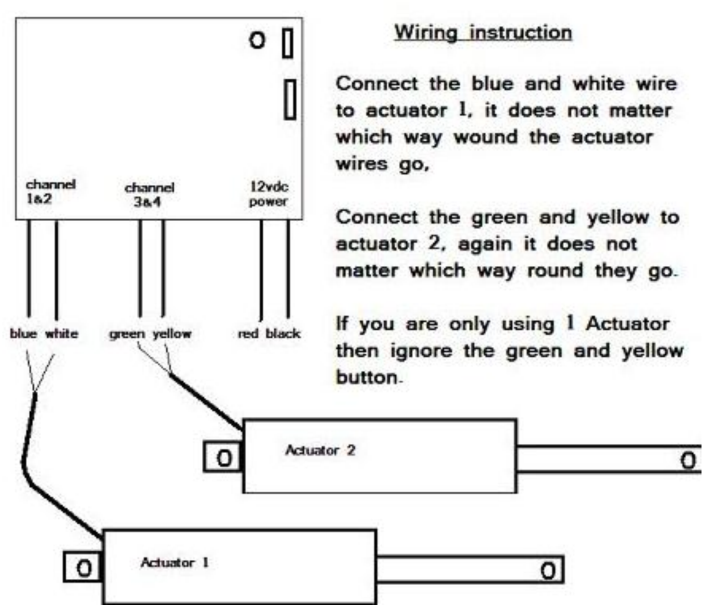


Figure 28. Camera video still frame of flame test taken through top viewing port

The senior project team HiTop succeeded in upgrading the HTTU by adding four new systems, which included viewing ports, a camera system, a leak detection system, and a flame impingement test. All four systems were successfully implemented, and all four showed functionality during testing. The system is still need refinement and further testing, which should be carried out by a future senior project team.

Figure 29. Completed HTTU Upgrade Project, as presented at the Senior Project Fair, May 30th 2013

Task 3. Improved Ceramic Filtration Materials

This was a productive year. Challenges were faced and overcome, and as a result, a number of inventions were discovered and corresponding patents filed. As such, Attachment A contains significant proprietary information on multiple aspects of ceramic filters.

Improvements in the individual filter element are necessary to maintain filtration efficiency while decreasing pressure drop. The path forward for an optimal ceramic HEPA filter meeting DOE's needs is to improve the filtration media. This task utilized the LLNL/DOE developed innovations along with progress in previous activities to prepare multiple ceramic filter elements and refine the technical details for depositing advanced filtration materials to the ceramic filter to meet desired filter performance. It is anticipated that the LLNL/DOE developed innovations provide an opportunity to reduce the pressure drop while maintaining filtration efficiency of DOE-STD-3020.

Research to improve filter media coating is being conducted to maintain filtration efficiency while improving performance (reducing pressure drop). A number of activities and efforts were involved with the filter media development; key activities were associated with (see Attachment A): 1) deposition of advanced materials as filter media, 2) an overwrap of inorganic filter media on porous ceramic tube substrates, and 3) the Flow Test System. The filter media fabrication laboratory was improved; a LLNL filter media deposition system to deposit advanced materials on a ceramic substrate has been designed, built, installed, completed safety requirements, and is in use. Details can be found in Attachment A. All the necessary safety reviews were successfully completed. The overwrap of inorganic filter media on porous ceramic tube substrates with the demonstration of HEPA filtration efficiency at acceptable flow rates and pressure drop before and after exposure to 500°C temperature is a key accomplishment in demonstrating a high temperature HEPA filter that exceeds the performance of existing filters in current use.

With regard to the development of nanofiber based filter media, extensive effort was devoted to developing methods and creating and depositing nanofibers (see Figures 30 and 31). Substantial efforts were made in developing processing techniques and parameters and in evaluating the conversion of the fibers to inorganic nanofibers. Various challenges were encountered and this led to use of several material systems to produce the nanofibers. These challenges, and solutions to overcome them, resulted in inventions for which patents have been filed (see Attachment A for additional information). The final selected precursor was a stabilized zirconium oxide composition. A technique was determined for depositing the filter media on to a small ceramic porous disk which was tested as a Small-scale Prototype Filter. This prototype filter demonstrated removal of approximately 75% or more of the Dioctyl Phthalate (DOP) challenge particles during testing. Improvements in the fabrication including sealing and filter media deposition are likely paths to obtaining improved performance that could approach the standard HEPA performance. A thicker layer of filter media would also be an approach to obtaining increased filtration efficiency. Overall, the test of the Small-scale Prototype gives an indication of the possibilities for a high temperature HEPA filter using the LLNL developed innovations.

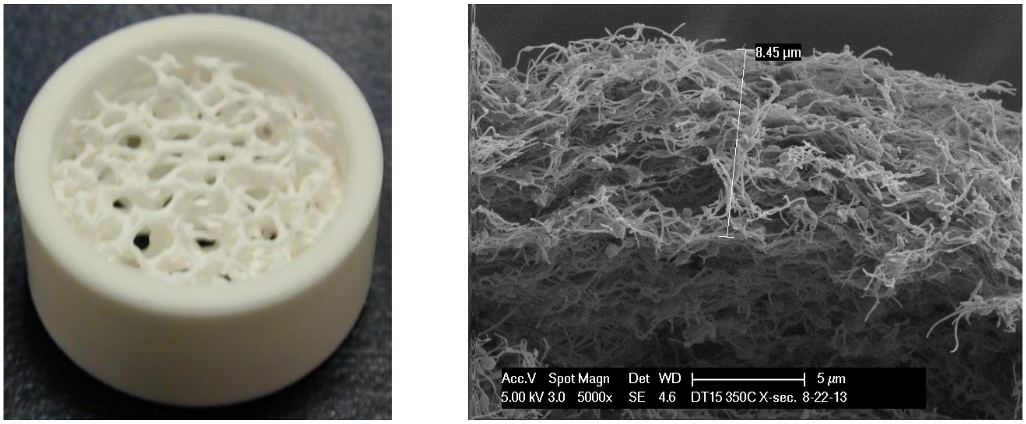
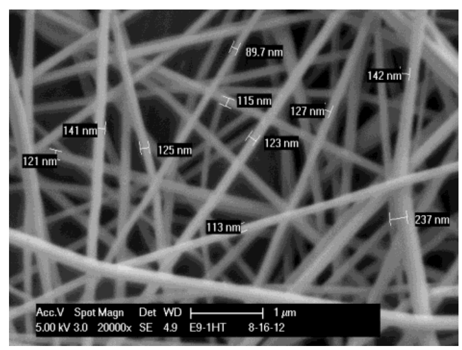


Figure 30. Nanofiber layers supported on porous ceramic framework

Silica

- Silica
  - 187nm ±31(95%) before HT
  - 124nm ±12 (95%) after HT
- ZrO_2 (~3m%Y_2O_3)
  - 47nm ±8 (95%) after HT



ZrO_2

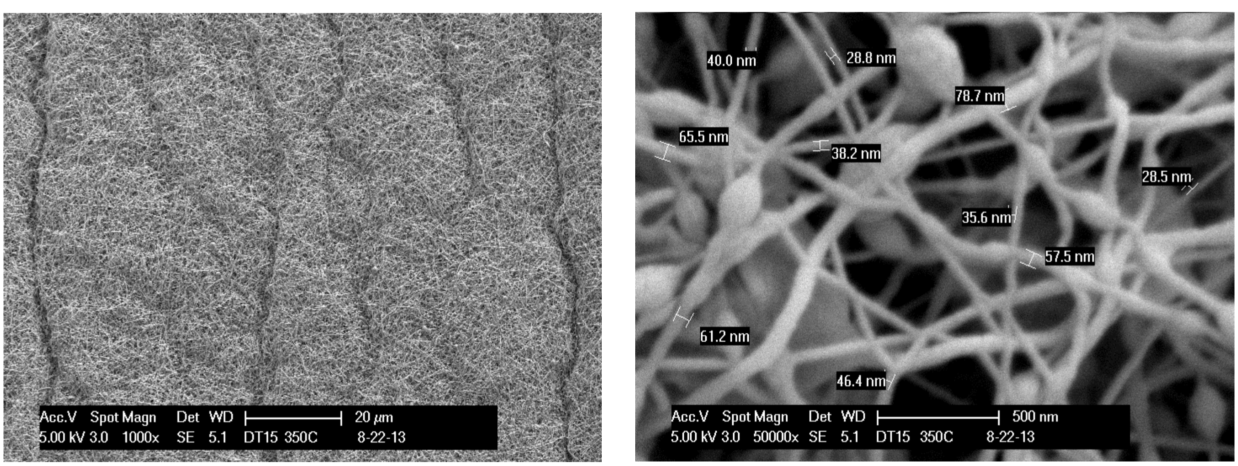


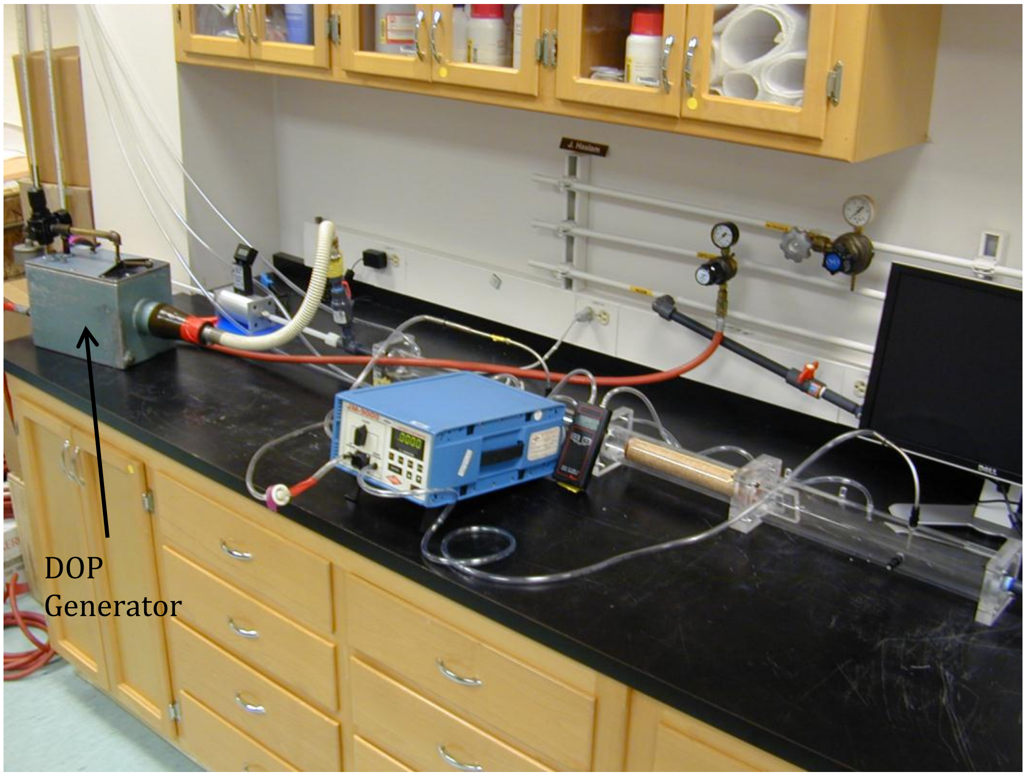
Figure 31. Nanofiber deposition

With regard to the development of tests of an overwrap of inorganic (ceramic/glass filter) media applied over a highly porous ceramic substrate such as tubular filter elements, a fabrication approach was developed and the filters were tested for pressure drop and filtration efficiency both before and after exposure to elevated temperature. The tube filters were developed using porous tube substrates and these filters utilized various inorganic filter media wrapped over the exterior of the porous tubes. A ceramic adhesive was used to seal the lap seam in the filter media. This filter media and tube filter combination demonstrated HEPA performance before and after heat treatment at 500°C for flow rates appropriate to the rated flow of a standard class of HEPA filter. A higher temperature filter media (vendor suggested use temperatures up to 1000°C) was also used to fabricate the ceramic tube filters. The high temperature filter media was successfully tested in the tube filter form after exposure to 250°C using a high temperature RTV to seal the lap seam. The performance in the higher temperature filter media was limited by the RTV. Additional effort will be needed to develop or determine an appropriate high temperature sealant for the higher temperature filter media. As a result of these efforts, we now have produced a proof of concept demonstration of an inorganic high temperature ceramic filter design that can meet the 99.97% removal of 0.3 micrometer particles specified for HEPA filters. This accomplishment will facilitate testing of this tube design, e.g., at ICET and the High Temperature Test Unit (HTTU).

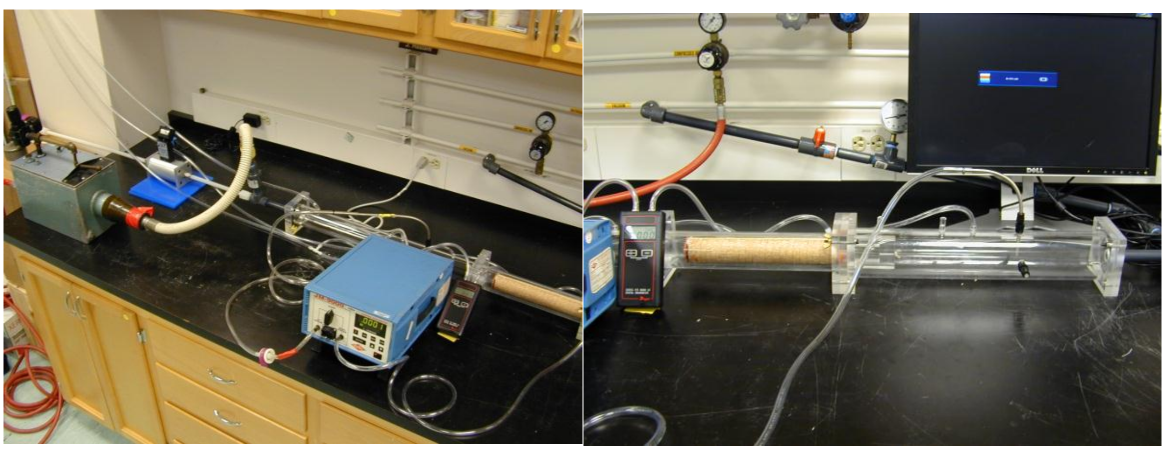
Figure 32. Picture of Tube Forms of Ceramic Filters with Lap Seams shown

The filter tube on the left has a spiral wrap crossing the ceramic cement lap seam in the filter media. The filter on the right has just the lap seam sealed by the ceramic cement.

The Flow Test System was successfully commissioned and utilized for testing both the ceramic tube filter and the Small-scale Prototype Filter as part of the third major activity for the project. A flow test apparatus was designed and built for testing the porous tube elements with the filter media applied to the exterior of the tube (see Figure 33). The system was capable of pressure and flow measurement along with particle removal efficiency measurement through the use of a DOP particle generation system and a photometer for measuring the particle concentration upstream and downstream of the filter. Tests were performed under conditions that should be representative of HEPA filter test conditions and nominally representative of conditions for a HEPA filter built as an assembly of the tubes in the standard classes of HEPA filter frames. The Flow Test System was useful for measuring the performance of tube filters (see Figure 32) at air flow rates up to about 10.5 cfm (300 lpm) on a single tube (cfm = cubic feet per minute, lpm = liters per minute). The Flow Test System has been commission and used extensively for testing and has been found to be quite valuable for testing filter elements during this year.



(a) Flow Test System including flow meter, DOP particle generator, Photometer (measures DOP particle concentration), pressure drop meter and upstream and downstream ducts to allow uniform flow and sampling of DOP particles



(b) Close up views of the upstream test equipment and the filter tube inside the flow test apparatus

Figure 33. Picture of Flow Test System

Brief Task 3 Deliverables Summary:

1. The Flow Test System was designed and commissioned and was used for testing of both filter tubes and the Small-scale Prototype Filter.
2. Pressure Drop tests at various appropriate flow rates and some higher than typical flow rates were reported for the tube filters and the Small-scale Prototype Filter
3. The nanofiber deposition system was assembled, commissioned, and used to perform deposition of nanofiber filter media.
4. Parameters were developed for the production of the nanofiber filter media.
5. Nanofibers were deposited and characterized including measurement of fiber diameter and some limited confirmation of the conversion to zirconium oxide using SEM and Energy Dispersive Spectroscopy (EDS) in the SEM.
6. Deposition parameters were refined for a polymer and an inorganic precursor system. The latest selected ceramic fiber system is a zirconium oxide precursor solution with yttrium oxide precursor added.
7. The filter tubes were tested utilizing the DOP particle generator and a photometer test system with the Flow Test System with measurements of pressure drop, flow rate, and filtration efficiency. Performance at 99.97% of 0.3 micron DOP particles was obtained from several tested tube filters.
8. The Small-scale Prototype was fabricated using the nanofiber filter media after conversion of the fibers to an oxide form (an LLNL/DOE developed innovation).
9. The Small-scale Prototype was tested for filtration efficiency and pressure drop using the Flow Test System with the DOP particle generator and Photometer measuring system.

Deliverable: Report on the project efforts and accomplishments (this document).

References:

1. LLNL-PROC-559284, *Ceramic HEPA Filter Program*, 32nd International Nuclear Air Cleaning Conference, Mark Mitchell et al., June 2012.

2. LLNL-CONF-554891, *Ceramic HEPA Filter Program*, EFCOG SAWG Safety Analysis Workshop, NSR&D Interest Group, Mark Mitchell et al., May 2012.

3. *High Temperature Test Unit: Control and Data Acquisition System*, Team CP presentation slides to LLNL, Matt Gainer, Marc Goupil, Andrew Woolrich, April 10, 2012.

4. *HEPA Filter Evaluation Furnace Control Unit Final Report*, Matt Gainer, Marc Goupil, Andrew Woolrich, [Team CP] Cal Poly, November 30, 2012.

5. *HTTU Upgrade Project,* HiTop presentation slides to LLNL, Blair Frandeen, Will Schill, Erick Shewmaker, Josh Turgeon., Cal Poly, February 8, 2013.

6. *Final Senior Project Report, High Temperature Filter Test Unit Upgrades,* Blair Frandeen, Will Schill, Erick Shewmaker, Josh Turgeon, [Team HiTop] Cal Poly, June 12, 2013.

7. *FY12 Summary Report for NSR&D Ceramic HEPA Filter Program, Improved Ceramic Filtration Materials Project*, J. Haslam et al, LLNL, October 2013. [

Attachment A. Improved Ceramic Filtration Material Report

Note: This attachment is Official Use Only and available separately. [5]

[5] This report is available in two versions: this summary document (which may be released to the public) with and without proprietary Attachment A (which cannot be released publically).